<u>Response to review comments</u>

acpd-15-20881-2015: Sensitivities of UK $PM_{2.5}$ concentrations to emissions reductions
Vieno et al.

We thank the reviewers for their time spent reviewing our manuscript and for highlighting the interesting and useful findings we report. Below we respond in turn to the additional individual comments made by the reviewers and indicate the revisions we have made to our paper. The original comments from the referees are in plain text and our responses are in *italic*.

Anonymous Referee #1

General: This study focuses on changes in $PM_{2.5}$ from emissions changes, but climate change itself may have an effect. Granted, the latter effect is usually found to be smaller, but it is probably worth mentioning the relevant lit on the topic

The focus of this work is an assessment of sensitivity of UK $PM_{2.5}$ surface concentrations to reductions of UK terrestrial emissions of relevant components/precursors. These reductions are with respect to a 'current' baseline and to a time-horizon relevant to developing and implementing additional policy on precursor emissions beyond those already in a 'current legislation' pipeline. 2030 was chosen as being not too soon for additional policy change to be implemented, but not too far that other factors might become overly influential (for example significant changes in global concentrations of methane, land-use change or climate). We agree that climate change may also have some effect on $PM_{2.5}$, but so might other factors (examples given above) which are not explicitly considered in this sensitivity study. In addition, as the reviewer notes, other work has shown that for the 2030 time horizon pollutant concentrations will be more strongly influenced by changes in pollutant emissions than by changes in climate (some citations given in the additional text below).

In response to the reviewer's comment, we have now added the following text to the revised paper: "It is recognised that climate change may also have some influence on future $PM_{2.5}$ concentrations in the UK; however the focus is here on UK precursor emission sensitivity and many studies have concluded that on the 2030 timescale air pollutant concentrations will be much more strongly influenced by changes in precursor emissions than by changes in climate (e.g. Langner et al. (2012);Colette et al. (2013);Coleman et al. (2013))."

20885.13: Please explain more the details of how observations are used to derive boundary conditions.
The import of $PM_{2.5}$ and its precursors to the UK from continental Europe is larger than for outside the European domain. The cross Atlantic transport of SIA has a small effect on the EU surface concentrations of SIA (as well as on deposition) as has been demonstrated by Simpson et al. (2012) and Sanderson et al. (2008). However, as European emissions are further reduced the relative cross Atlantic transport may play a larger role in the future

We have added the following text to the manuscript:
"The EMEP4UK model uses a nested approach, the European domain concentrations are used as boundary condition for the UK domain. The boundary condition at the edge of the European domain are prescribed concentrations in terms of latitude and adjusted for each year. For ozone, 3-D fields for the whole domain are specified from climatological ozone-sonde data-sets, modified monthly against clean-air surface observations as described in Simpson et al. (2012)"
.

20885.20: What is the potential limitation? Can it be briefly described? At present this statement is too vague to be informative

The following text is now included in the revised paper in place of the original sentence that read "The potential limitation of the EMEP4UK approach is discussed in (Vieno et al., 2014)"

"Whilst fine nitrate production is modelled using a thermodynamic model (MARS), the formation of coarse nitrate from nitric acid uses a parameterised approach that seeks to capture the HNO_3 *reaction with sea salt and crustal material. The conversion rate of* HNO_3 *to coarse nitrate depends on relative humidity, as described by Simpson et al. (2012), but is not explicitly linked to the surface area of the existing coarse aerosol. Both nitrate generation mechanisms compete for the same* HNO_3*, and whilst this constrains the total amount of nitrate produced, it is acknowledged that the resulting split into fine and coarse nitrate is somewhat uncertain as discussed in Aas et al. (2012). A more explicit aerosol scheme is under development for the model."*

Moreover, we apologise that there was an error in the original text in that the version of EMEP4UK used in this study used the MARS rather than the EQSAM aerosol scheme. We have now corrected the text and amended the associated reference.

20887.1: How realistic is it to assume the spatial distribution is fixed? How might this bias the results of this work?

We agree that the spatial distribution of emissions is likely to change with time; for example, point source emissions can change between years (e.g. power plant closing or new build). Land-use changes may also affect ammonia emissions. However, these changes are not easily predicted for a future scenario and it might be anticipated that changes in spatial patterns of emissions will likely be smaller than the changes in absolute amounts of emissions. We have added the following sentence to acknowledge this point in the text:
"Whilst there will likely be some changes in the spatial distribution of emissions, such changes are not easily predicted for a future scenario, and are anticipated to be smaller than the changes in absolute amounts of emissions."

In addition, the focus of this study is on quantifying the contribution of different reduction options, so while changes in distribution patterns will affect comparisons between modelled and observed surface concentrations in the future, the quantification of the relative effect of emission reductions as a UK average should be relatively less affected.

20887.5: As mentioned above, some discussion of how large such impacts might be would be worth including

We already mention in the Discussion that inter-annual variability in meteorology will mean that the sensitivities of UK $PM_{2.5}$ *to additional reductions in a variety of precursor emissions will also have some inter-annual variability. However, as emphasised above and in the paper, the focus of this work was on understanding the effect of these emissions sensitivities which was why meteorology was deliberately kept constant to isolate these. Whilst it is possible that climate change by 2030 may also change the nature of the UK meteorology (to a small extent), as indicated in one of our responses above, any such impact on this time horizon is likely to be much smaller than the impact of precursor emissions changes.*

20887.23: Can the authors be more rigorous here and provide statistical analysis such as correlation coefficients, bias and error.

We have now added the linear regression between observation and model at the top of each panel, along with the correlation coefficient, bias, and mean square error, and also a summary of the comparison reported in Vieno et al. (2014). The four sites included in Vieno et al. 2014 showed good agreement between EMEP4UK simulation and the observed NO_3^- and SO_4^{2-} as shown in the Table 1 of this paper. This is reproduced below for these responses but not in the main paper since this provides the citation to the original source.

*Table 1: Mean concentrations, and correlation and regression statistics, for monthly-averaged modelled and measured NO_3^- and SO_4^{2-} in particulate matter for the period 2001-2010 at four sites of the AGANet network: Strathvaich Dam (north-west Scotland), Bush 1 (central Scotland), Rothamsted (south-east England), and Yarner Wood (south-west England). The comparison is based on a linear fit where measurement = slope * model + intercept.*

	Particulate NO_3^-			
	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
Measurement mean	0.49 μg m^{-3}	1.37 μg m^{-3}	3.35 μg m^{-3}	1.98 μg m^{-3}
Model mean	0.77 μg m^{-3}	1.42 μg m^{-3}	2.73 μg m^{-3}	2.23 μg m^{-3}
R	0.49	0.91	0.81	0.86
Slope	0.59	0.96	0.68	0.95
Intercept	0.48 μg m^{-3}	0.10 μg m^{-3}	0.44 μg m^{-3}	0.34 μg m^{-3}
	Particulate SO_4^{2-}			
	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
Measurement mean	0.57 μg m^{-3}	0.94 μg m^{-3}	1.75 μg m^{-3}	1.20 μg m^{-3}
Model mean	0.61 μg m^{-3}	0.95 μg m^{-3}	1.48 μg m^{-3}	1.28 μg m^{-3}
r	0.72	0.79	0.65	0.69
Slope	0.86	0.76	0.56	0.65
Intercept	0.12 μg m^{-3}	0.24 μg m^{-3}	0.50 μg m^{-3}	0.36 μg m^{-3}

20887.24: Can the authors summarize the species specific evaluation of Conolloy 2011? Does the model do better at estimating concentrations of any particular component of SIA? What were the biases, quantitatively?

The citation to Conolly et al. (2011) is for the description of the monitoring network; the model-observation evaluations against the monitor data were presented in Vieno et al. (2014), but as requested in the previous comment we have now added the essential aspects of the evaluation data to this paper too. We have also added a reference to another study in which the EMEP4UK model was evaluated against observations and other model (Carslaw, 2011a, b).

20881.1: This seems rather unsubstantiated. How did the authors rule out the role of SOA? Why was particle-bound water not included in the model PM2.5 calculations (it is easily done using hydroscopic growth curves â˘A˘T textbook undergraduate level calculation)?

We apologise in not being explicit in the model description section in stating that the EMEP4UK model does include both primary organic aerosols and secondary organic aerosols. We have now added the following description of this to the revised paper:

“In the model version used here, $PM_{2.5}$ is the sum of the fine ($PM_{2.5}$) fraction of: ammonium (NH_4^+), sulphate (SO_4^{2-}), nitrate (NO_3^-), elemental carbon (EC), organic matter (OM), sea salt (SS), mineral dust, and 27% of the coarse nitrate. PM_{10} is the sum of $PM_{2.5}$ plus the coarse ($PM_{2.5-10}$) fraction of EC, OM, NO_3^-, SS, and dust.”
The reason why the paper does not discuss SOA is because the impact of reductions in emissions of UK VOC has very little impact (via formation of anthropogenic SOA) on the UK $PM_{2.5}$ as shown in Figures 4e (or 2010 scenario) and 8e (for the 2030 scenario), as compared with the impact on $PM_{2.5}$ for emissions reductions in primary $PM_{2.5}$, NH_3, SO_2 and NO_x. (Note that the impact of primary $PM_{2.5}$ emissions reductions on UK $PM_{2.5}$ will include the contribution from reductions in primary organic aerosol.)

The issue of whether or not to include an estimate of particle-bound water is not straightforward. Different measurement techniques and conditions will incorporate different proportions of the ambient $PM_{2.5}$ water content. Because of this uncertainty in what measurements measure (against which legislation for PM is based), we focus here on changes to the dry mass of surface $PM_{2.5}$ derived from changes in the emissions of primary $PM_{2.5}$ and in secondary $PM_{2.5}$ precursor gases. However, we acknowledge that changes in mass of secondary inorganic components will be accompanied by changes in mass of particle-bound water and now incorporate caveats to this effect in the both the Results and Discussion section .

20888.2: What evidence do the authors provide that such missing mechanism don’t affect the sensitivities calculated here? It’s not entirely implausible. For example, if they have neglected uptake of HNO_3 on dust, then they are overestimating their response of nitrate to changes in NO_x emissions…Or if they considered the role of NO_x on SOA, which can be quite significant.

The RH-dependent coarse nitrate formation parameterises the effect of HNO_3 onto mineral dust, however the details are not reproduced mechanistically (see response 20885.20). A new version of the EMEP4UK model is currently under development that will explicitly calculate this but is not available at this time. We confirm again, however, that the model does represent the changes in SOA due to emissions reduction in NO_x (and other relevant chemistry). Description stating that SOA is included in the model has been added to the Methods section.

20888.8: Likewise, some overestimation would occur for the background site. This affect may somewhat cancel the low-biases discussed above.

For rural background sites without significant emissions the grid average should be representative, rather than overestimated. However, the two rural background AURN sites which have enough data to compare the monthly values are Harwell and Auchencorth. At this two sites the bias is -1.1 and 2.5 (µg m^{-3}) for Harwell and Auchencorth Moss, respectively.

20888.10 & 26: It seems like one additional model run with boundary conditions set to zero could easily be performed to quantify this aspect more completely.

This has undertaken and presented previously in an report of the Air Quality Expert Group (AQEG, 2015) The analysis showed that UK emissions contribute around 50-55% of total annual average $PM_{2.5}$ in the UK. We add the following text and reference in the manuscript:
“An analysis presented in AQEG (2015) also using the EMEP4UK model showed that UK emissions contribute around 55% of the total $PM_{2.5}$ in the UK. This limits the extent to which long-term average concentrations can be reduced by UK action alone”

20889.13: This results is “key” to what, exactly? Also, why does this occur, from a standpoint of atmospheric chemistry and aerosol partitioning? If the authors wish to draw attention to this finding, it should be better explained.

The word ‘key’ here has now been replaced by ‘important’. The result to which this comment refers is the observation of different spatial patterns for reductions in $PM_{2.5}$ *across the UK for different precursor reductions. Rationalisation for these different patterns is discussed at a number of places in the Results and Discussions sections (e.g. the different locations of the sources of different precursors and the different timescales, and hence spatial scales, over which chemistry and transport interact).*

20889.22: why?

This query is referring to our statement that reductions in $PM_{2.5}$ *arising from reductions in* SO_x *emissions are not generally associated with urban areas. This is because the major sources of* SO_x *emissions in the UK are large power-plants and large industrial plants (e.g. steelworks) that are not located within urban areas but outside urban areas. The couple of subsequent sentences we already have in the text provide rationalisation, but for further emphasis we have now added the following sentence after the one in the original text: “This is primarily caused by the spatial distribution of major sources of* SO_x *emissions. As ~80% of UK* SO_x *2010 emissions originate from large point sources (power plants, industrial facilities), which are not located in the heart of urban areas, associated emission reductions have the most profound effects in rural areas.”*

20890.1: This could have been determined without any sensitivity model experiments by calculating the gas ratio (= available NH3 beyond that required for sulfate neutralization, divided by total inorganic nitrate + nitric acid, Ansari and Pandis, 1998) in the baseline model run. A map of that ratio over the UK would be useful for this work.

We agree that the NH_3 *sensitivity of* $PM_{2.5}$ *formation could have been demonstrated by the method described, but we have demonstrated it as a consequence of the emissions sensitivities runs performed for this work. Moreover, it is not so straightforward since changes in emissions also change the dry/wet deposition ratio which also affects the lifetime and hence concentrations of* $PM_{2.5}$ *(Vieno et al., 2010).*

20892.17: To be fair though, it may also be worth mentioning that NH_3 reductions would as well have additional benefits given their impacts on N deposition and ecosystems.

We entirely agree that NH_3 *emissions reductions (Klimont and Winiwarter, 2015), and reductions in other precursors, will have other additional benefits on, for example N (and S) deposition (Adrian et al., 2015) and on ground-level ozone (with its associate human health and ecosystem impacts), aside from the impacts of these emission reductions on* $PM_{2.5}$ *that are the focus of this work. Our paper already pointed this out in a number of places. For example, right at the outset, at the end of the Introduction we state “It is also recognised that, whilst the focus here is on reduction in concentrations of* $PM_{2.5}$ *from the perspective of its impact on human health, the reduction of anthropogenic emissions in general will also have other benefits including on human health, on N and S deposition, and on ozone formation”; in the second paragraph of the discussion we write: “It is also recognised that reductions in* NO_x *and VOC emissions have the potential to deliver health benefits separately from their contribution to reduction in* $PM_{2.5}$ *through reductions in population exposure to surface* NO_2 *and* O_3*”; and at the end of the Discussion we write: “ Measures taken in the UK to reduce concentrations of ambient* $PM_{2.5}$ *and of precursor gases, both within and outside of areas of population, will have multiple co-benefits on human health, N and S deposition, ozone formation and radiative forcing, not* *just in the UK but elsewhere." We therefore think we have adequately emphasised the other benefits of emissions reductions.*

MINOR COMMENTS:
20882.18: "but" – what is being contrasted here? Maybe a different word would be better.

The word 'but' has been changed to 'however'.

20882.24: "observation" – conclusion? "observation" implies a measurement

The word 'observation' has been changed to 'conclusion'.

20883.9: should clarify that 3.2 million is exposure to ambient PM2.5 (as opposed to indoor, which is even larger)

The word 'ambient' has now been inserted to read '..to exposure to ambient PM2.5 concentrations prevailing in 2005…'

20884.1: I don't see how the mass concentrations masks composition. This could be re-written to be better, something like "while standards focus on PM2.5 mass concentration, meeting these standards are complicated by the considerable chemical heterogeneity. . ."

We agree the original phrasing was probably a bit difficult to interpret to understand what we meant. The reviewer supplies a good alternative phrasing to the start of this sentence which we have now incorporated.

20884.21: usually write old to new in citations years

Multiple in-text citations are now listed in chronological order of publication.

20885.16: reactions,

A comma has now been inserted after 'reactions' in this sentence.

20888.16: It would be interesting to also see the % reductions.

Throughout this work we focus on the absolute reductions in $PM_{2.5}$ (i.e. $\mu g\ m^{-3}$) derived from the UK emissions reductions simulations because it is the change in absolute amount of $PM_{2.5}$ that drives the change in impact of $PM_{2.5}$ (on health, or deposition, etc.) and because legislation for compliance on $PM_{2.5}$ concentrations is quantified by the amount of $PM_{2.5}$. Our work is an exploration, from the 'policy-maker perspective,' of sensitivities of UK $PM_{2.5}$ reductions to UK emissions reductions and it is the resultant absolute changes in $PM_{2.5}$ that are of relevance to policy-makers (and to the $PM_{2.5}$ impacts).

Reviewer T. Oxley

General Comments: This is a useful paper describing the sensitivity of UK PM2.5 concentrations and therefore health impacts to changes in primary and secondary pre-cursor emissions using the EMEP4UK ACTM.

We thank the reviewer for their supportive comments on the work.

I found that figure 6 not only doesn't add to the manuscript, but that it actually made it more difficult to follow because it took me some time to work out what the map was actually showing me, ie a 30% reduction in NH3 combined with a 30% increase in pPM2.5, which was a bit strange? The point the authors make regarding urban or rural impacts is valid, but I had already understood this from Figure 4.

We believe that Figure 6 is an impotant figure for this paper in that it provides a direct visualisation of the spatial pattern across the UK of localities where $PM_{2.5}$ reductions are most effectively derived through primary $PM_{2.5}$ emissions reductions as compared with through NH_3 emissions reductions (on the basis of applying 30% emissions reductions to one or the other). In this single visualisation, Figure 6 directly shows that for the largest urban areas in the UK, reducing primary $PM_{2.5}$ emissions is more effective at reducing $PM_{2.5}$ than the equivalent percentage emissions reductions in NH_3; whereas outside these urban areas, reducing NH_3 emissions is more effective at reducing $PM_{2.5}$ than the equivalent percentage emissions reduction in primary $PM_{2.5}$.

We think the caption text for this figure is clear in stating how this figure is derived and how it should be interpreted: first, the caption explicitly states that Figure 6a is the data in Figure 4b ($PM_{2.5}$ changes for 30% NH3 emissions reductions) minus the data in Figure 4f ($PM_{2.5}$ changes for 30% primary $PM_{2.5}$ emissions reductions) for the 2010 year, and similarly for Figure 6b for the 2030 future scenario; secondly, the caption also states that blue colours indicate where reductions in $PM_{2.5}$ for 30% reduction in NH3 emissions exceed the reductions in $PM_{2.5}$ for 30% reduction in primary $PM_{2.5}$ emissions, and vice versa for the red colours.

We therefore wish to retain Figure 6. It may be that the short title that was also present on this figure contributed to some misunderstanding in interpretation; we have now removed this title so that the caption alone provides the detail.

Specific Comments:
Is figure 2 really necessary as it is simply a population map which could be downloaded from www.ons.gov.uk

A central finding of our work is the different effects on $PM_{2.5}$ mitigation spatially that derive from UK emissions reductions of different $PM_{2.5}$ precursors, which in turn means that different precursor emissions reductions have different impact on population-weighted $PM_{2.5}$ compared with area-weighted $PM_{2.5}$. Understanding this point is helped by knowledge of the pattern of population density in the UK. Whilst we could just retain the summary data for the effects of the reductions on the UK national population-weighted and area-weighted $PM_{2.5}$ that appear in Table 1 and Figure 5, we believe that since our paper includes a number of maps of model simulations it is helpful for the reader to be able to see directly for themselves the geographical comparison between maps of spatial changes in $PM_{2.5}$ for different emissions reductions sensitivities and maps of the population density of the UK. We think this is particularly important for readers not familiar with the UK urban layout. We note this reviewer is UK-based so will presumably be very familiar with the locations of UK urban centres. This will not be familiar to many other readers.

Whilst we could just refer the reader to a URL, or put the population weighted map in supplementary information, since we are talking about one figure that we believe will be of considerable help to the reader in emphasising our findings we wish to retain this Figure in the main paper.

P20890, L14: I think it would benefit to remove Figure 6 and revise these paragraphs accordingly so that the discussion of 2010 and 2030 effects flows better. P20891, L1: Figure 8 I like. Figure 6b is definitely unnecessary having seen figure 8.

These two comments refer again to the issue of whether to retain Figure 6. See our response to this comment above.

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The sensitivities of emissions reductions for the mitigation of UK $PM_{2.5}$

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ABSTRACT

The reduction of ambient concentrations of fine particulate matter ($PM_{2.5}$) is a key objective for air pollution control policies in the UK and elsewhere. Long-term exposure to $PM_{2.5}$ has been identified as a major contributor to adverse human health effects in epidemiological studies and underpins ambient $PM_{2.5}$ legislation. As a range of emission sources and atmospheric chemistry transport processes contribute to $PM_{2.5}$ concentrations, atmospheric chemistry transport models are an essential tool to assess emissions control effectiveness. The EMEP4UK atmospheric chemistry transport model was used to investigate the impact of reductions in UK anthropogenic emissions of primary $PM_{2.5}$, NH_3, NO_x, SO_x or non-methane VOC on surface concentrations of $PM_{2.5}$ in the UK for a recent year (2010) and for a future current legislation emission scenario (2030). In general, the sensitivity to UK mitigation is rather small. A 30% reduction in UK emissions of any one of the above components yields (for the 2010 simulation) a maximum reduction in $PM_{2.5}$ in any given location of ~0.6 µg m^{-3} (equivalent to ~6% of the modelled $PM_{2.5}$). On average across the UK, the sensitivity of $PM_{2.5}$ concentrations to a 30% reduction in UK emissions of individual contributing components, for both the 2010 and 2030 CLE baselines, increases in the order NMVOC, NO_x, SO_x, NH_3 and primary $PM_{2.5}$; however there are strong spatial differences in the $PM_{2.5}$ sensitivities across the UK. Consequently, the sensitivity of $PM_{2.5}$ to individual component emissions reductions varies between area and population weighting. Reductions in NH_3 have the greatest effect on area-weighted $PM_{2.5}$. A

full UK population weighting places greater emphasis on reductions of primary $PM_{2.5}$ emissions, which is simulated to be the most effective single-component control on $PM_{2.5}$ for the 2030 scenario. An important conclusion is that weighting corresponding to the Average Exposure Indicator metric (using data from the 45 model grids containing a monitor whose measurements are used to calculate the UK AEI) further increases the emphasis on the effectiveness of primary $PM_{2.5}$ emissions reductions (and of NO_x emissions reductions) relative to the effectiveness of NH_3 emissions reductions. Reductions in primary $PM_{2.5}$ have the largest impact on the AEI in both 2010 and the 2030 CLE scenario. The summation of the modelled reductions to the UK $PM_{2.5}$ AEI from 30% reductions in UK emissions of primary $PM_{2.5}$, NH_3, SO_x, NO_x and VOC totals 1.17 μg m^{-3} and 0.82 μg m^{-3} for the 2010 and 2030 CLE simulations, respectively.

1 Introduction

Atmospheric particulate matter (PM) has a range of adverse impacts including on climate change through radiative forcing (IPCC, 2013) and on human health (WHO, 2006, 2013). The global health burden from exposure to ground-level ambient fine particulate matter (as characterised by the $PM_{2.5}$ metric) is substantial. The Global Burden of Disease project attributed 3.2 million premature deaths and 76 million disability-adjusted life years to exposure to ambient $PM_{2.5}$ concentrations prevailing in 2005 (Lim et al., 2012). Exposure to ambient $PM_{2.5}$ remains a major health issue in Europe. The European Environment Agency report that for the period 2010-2012, 10-14% of the urban population in the EU28 countries was exposed to ambient concentrations of $PM_{2.5}$ exceeding the EU annual-average $PM_{2.5}$ reference value of 25 μg m^{-3}, but 91-93% were exposed to concentrations exceeding the WHO annual-average $PM_{2.5}$ air quality guideline of 10 μg m^{-3} (EEA, 2014).

European Commission (EC) legislation for $PM_{2.5}$ includes an obligation on individual member states to reduce exposure to $PM_{2.5}$ in areas of population by a proscribed percentage between 2010 and 2020. The exposure to $PM_{2.5}$ is quantified through the Average Exposure Indicator (AEI) which is the average of the annual $PM_{2.5}$ measured across designated urban background and suburban sites spread over cities and large towns (averaged over the 3-year periods spanning 2010 and 2020). The AEI is therefore a quasi-indicator of population-weighted $PM_{2.5}$.

For the UK, the calculation of the AEI uses data from 45 sites (Brookes et al., 2012) and the required reduction by 2020 is 15% from its 2010 value of 13 μg m^{-3} (Defra, 2012).

While standards focus on $PM_{2.5}$ mass concentration, meeting these standards is complicated by the considerable chemical heterogeneity, which arises because ambient $PM_{2.5}$ comprises both primary PM emissions and secondary inorganic and organic components formed within the atmosphere from gaseous precursor emissions, specifically NH_3, NO_x (NO & NO_2), SO_2 and a wide range of non-methane volatile organic compounds (VOC) (USEPA, 2009; AQEG, 2012). Meteorological conditions also control $PM_{2.5}$ concentrations through their influences on dispersion, chemistry and deposition.

European legislation sets current and future caps on anthropogenic emissions of primary and secondary-precursor components of $PM_{2.5}$ at national level and from individual sources (Heal et al., 2012). Although it is well-known that much of the ambient $PM_{2.5}$ in the UK derives from trans-boundary emissions and transport into the UK (Vieno et al., 2014;AQEG, 2015), a pertinent policy question to address is: what additional surface $PM_{2.5}$ reductions could the UK unilaterally achieve, at least in principle? In other words, what are the sensitivities of UK $PM_{2.5}$ to UK reductions in emissions of relevant components?

This is the motivation for the work presented here, which investigates the impact of reductions from UK anthropogenic sources of emissions of primary $PM_{2.5}$ and of precursors of secondary $PM_{2.5}$ on surface $PM_{2.5}$ concentrations across the whole UK. To adequately simulate the UK national domain requires the use of a regional atmospheric chemistry transport model (ACTM), in this study the EMEP4UK Eulerian ACTM (Vieno et al., 2009;Vieno et al., 2010;Vieno et al., 2014). Recognising that reductions in UK and rest-of-Europe emissions are already projected under current legislation, this work compares the present-day sensitivity of UK emissions reductions on UK $PM_{2.5}$ with a future time point (2030) to examine the effectiveness of potential options in the future. It is recognised that climate change may also have some influence on future $PM_{2.5}$ concentrations in the UK; however the focus is here on UK precursor emission sensitivity and many studies have concluded that on the 2030 timescale air pollutant concentrations will be much more strongly influenced by changes in precursor emissions than by changes in climate (e.g. Langner et al. (2012);Coleman et al. (2013);Colette et al. (2013)).

Throughout, the focus is on annual average $PM_{2.5}$, since this is the metric within the AEI, which in turn is driven by the evidence from epidemiological studies that demonstrate associations between adverse health outcomes and long-term (annual average) concentrations of $PM_{2.5}$ (COMEAP, 2010;WHO, 2013). It is also recognised that, whilst the focus here is on reduction in concentrations of $PM_{2.5}$ from the perspective of its impact on human health, the reduction of anthropogenic emissions in general will also have other benefits including on human health, on N and S deposition, and on ozone formation.

2 Methods

2.1 Model description and set-up

The EMEP4UK model used here is a regional ACTM based on version rv4.4 (www.emep.int) of the EMEP MSC-W model which is described in Simpson et al. (2012). A detailed description of the EMEP4UK model is given in Vieno et al. (2010), and Vieno et al. (2014).

The EMEP4UK model meteorological driver is the Weather Research and Forecast (WRF) model version 3.1.1 (www.wrf-model.org). The EMEP4UK and WRF model horizontal resolution is 50 km × 50 km for the extended European domain and 5 km × 5 km for the inner domain as illustrated in Figure 1. The EMEP4UK model uses a nested approach, the European domain concentrations are used as boundary condition for the UK domain. The boundary conditions at the edge of the European domain are prescribed concentrations in terms of latitude and adjusted for each year. For ozone, 3-D fields for the whole domain are specified from climatological ozone-sonde data-sets, modified monthly against clean-air surface observations as described in Simpson et al. (2012).

The default EMEP MSC-W chemical scheme was used for the present study, as it has been extensively validated at the European scale (Simpson et al. (2012), www.emep.int). The scheme has 72 species and 137 reactions, and full details are given in Simpson et al. (2012). The gas/aerosol partitioning is the model for aerosols reacting system (MARS) formulation (Simpson et al., 2012). In the model version used here, $PM_{2.5}$ is the sum of the fine ($PM_{2.5}$) fraction of: ammonium (NH_4^+), sulphate (SO_4^{2-}), nitrate (NO_3^-), elemental carbon (EC), organic matter (OM), sea salt (SS), mineral dust, and 27% of the coarse nitrate. PM_{10} is the sum of $PM_{2.5}$ plus the coarse ($PM_{2.5-10}$) fraction of EC, OM, NO_3^-, SS, and dust.

Whilst fine nitrate production is modelled using a thermodynamic model (MARS), the formation of coarse nitrate from nitric acid (HNO_3) uses a parameterised approach that seeks to capture the HNO_3 reaction with sea salt and crustal material. The conversion rate of HNO_3 to coarse nitrate depends on relative humidity, as described bySimpson et al. (2012), but is not explicitly linked to the surface area of the existing coarse aerosol. Both nitrate generation mechanisms compete for the same HNO_3, and whilst this constrains the total amount of nitrate produced, it is acknowledged that the resulting split into fine and coarse nitrate is somewhat uncertain as discussed in Aas et al. (2012). A more explicit aerosol scheme is under development for the model

Anthropogenic emissions of NO_x, NH_3, SO_2, primary $PM_{2.5}$, primary PM_{coarse}, CO and non-methane VOC for the UK are derived from the National Atmospheric Emission Inventory (NAEI, http://naei.defra.gov.uk) at 1 km × 1 km resolution and aggregated to 5 km × 5 km resolution. For the European domain, the model uses the EMEP 50 km × 50 km resolution emission estimates provided by the Centre for Emission Inventories and Projections (CEIP, http://www.ceip.at/). Shipping emissions estimates, for the inner domain, are derived from the ENTEC (now Amec Foster Wheeler) emissions estimate (ENTEC, 2010). Natural emissions of isoprene and DMS are as described in.

The EMEP MSC-W model from which the EMEP4UK model is derived is used widely in support of European air quality science and policy development and the performances of both have been extensively evaluated (Carslaw, 2011b;Schulz et al., 2013;Simpson et al., 2014;Schaap et al., 2015).

2.2 Model experiments

A base run and a set of 5 sensitivity experiments were carried out for emissions and meteorology for 2010. The experiments applied 30% reductions to UK anthropogenic emissions from all sectors for each of the following pollutants individually: primary $PM_{2.5}$, NH_3, NO_x, SO_x and NMVOC. This 30% perturbation was applied to land-based emissions only; shipping emissions (both domestic and international) were left unchanged.

Model runs were repeated for a 2030 future emissions scenario to investigate sensitivities of UK $PM_{2.5}$ to UK emissions reductions further along the pathway of current legislation (CLE) emissions. The 2030 CLE emissions used in the model runs were based on the 2030 IIASA CLE projection (IIASA, 2012) for Europe and the *Updated Energy Projections* (UEP, version 45) for the UK. The UEPs are developed and regularly updated by analysing and projecting future energy use and are based on assumptions of future economic growth, fossil fuel prices, UK population development and other key variables. A set of projections is based on a range of assumptions to represent the uncertainty in making such projections into the future. For this manuscript, the mid-range estimates were used. For a full description of the UEPs and the methodology for their compilation, see DECC (2015). Emissions from shipping were 2020 emissions estimate provided by ENTEC (now Amec Foster Wheeler) (ENTEC, 2010).

No change in the spatial distribution of emissions was made. Whilst there will likely be some changes in the spatial distribution of emissions, such changes are not easily predicted for a future scenario, and may be anticipated to be smaller than the changes in absolute amounts of emissions. The boundary and initial conditions for ozone and particles outside the European domain were left unchanged to the year 2010, as was the meteorology. The use of the same meteorology isolates the sensitivity of surface $PM_{2.5}$ to emissions reductions at some future date from the effects on surface $PM_{2.5}$ due to differences in meteorology.

As well as maps of annual-average surface $PM_{2.5}$ concentrations the following three summary statistics for UK $PM_{2.5}$ were calculated: (i) the area-weighted average, i.e. the average of all 5 km × 5 km model grids over the UK; (ii) the population-weighted average, i.e. the 5 km × 5 km gridded estimates of $PM_{2.5}$ surface concentrations re-projected onto the British National Grid and multiplied by population estimates at the same spatial resolution (derived from the UK census, http://census.edina.ac.uk/) (Figure 2) and divided by the sum of the UK population; (iii) a value analogous to the Average Exposure Indicator (AEI), calculated as the average of the concentrations for the 45 model grids containing a $PM_{2.5}$ monitor whose measurements are used to define the UK's 2010 AEI value (Brookes et al., 2012).

3 Results

Example comparisons between EMEP4UK-modelled surface concentrations of $PM_{2.5}$ components and total measured $PM_{2.5}$ are shown in Figure 3 for three UK national network monitoring sites: Edinburgh St. Leonards, an urban background site in the north of the UK; London North Kensington, an urban background site in central London in the south-east of the UK; and Harwell, a rural background site in central England. Monthly averages of the hourly measured and modelled data are presented. Model simulations follow the observational time trends well. The model simulations of the SIA components SO_4^{2-}, NO_3^{-} and NH_4^{+} have previously been individually evaluated by Vieno et al. (2014) against 10 years of speciated observations made at ~30 sites across the UK in the AGANET network (Conolly et al., 2011). The four UK sites included in Vieno et al. (2014) showed good agreement between the EMEP4UK simulation and the observed NO_3^{-} and SO_4^{2-} , with a bias range of 0.28 to -0.62 and 0.8 to -0.27 $\mu g\ m^{-3}$, respectively. The EMEP4UK model was also evaluated against observations and other models in a UK model inter-comparison organised by the UK Department for Environment, Food & Rural Affairs (Defra) (Carslaw, 2011b, a). The persistent negative bias in the sum of the modelled $PM_{2.5}$ against observation in Figure 3 is consistent with the absence of re-suspended dust in the model configuration used here, and possibly also reflects a difference in the treatment of particle-bound water in model and measurement. The omission of re-suspended dust does not impact on the investigations here of the sensitivities of $PM_{2.5}$ concentrations to anthropogenic emissions reductions; however it is acknowledged that since particle-bound water is related to the mass of secondary inorganic components its omission will have some impact on the sensitivity of $PM_{2.5}$ to inorganic precursor gas emissions reductions. Different measurement techniques and conditions incorporate different proportions of the ambient $PM_{2.5}$ water content. Because of uncertainty in what measurements measure (against which legislation for PM is based), we focus here on changes to the dry mass of surface $PM_{2.5}$ derived from changes in the emissions of primary $PM_{2.5}$ and in secondary $PM_{2.5}$ precursor gases. (It is also noted that values of relative reductions in modelled $PM_{2.5}$ will be slightly higher than if expressed relative to measured $PM_{2.5}$ at that location.) Some model underestimation may also derive from dilution of primary $PM_{2.5}$ emissions into the 5 km grid of the model compared with the primary emissions more local to an urban background monitor.

The simulated 'baseline' 2010 annual-average surface concentrations for $PM_{2.5}$ at 50 km horizontal resolution for the EMEP4UK European domain and for the nested 5 km horizontal resolution British Isles domain are shown in Figure 1. The UK 2010 annual-average surface concentrations of $PM_{2.5}$ are generally lower compared with neighbouring continental countries such as France, the Netherlands and Germany. The influence of emissions originating from continental Europe is revealed by the gradient of decreasing $PM_{2.5}$ concentrations away from the continent. An analysis presented in AQEG (2015) also using the EMEP4UK model showed that UK emissions contribute around 55% of the total $PM_{2.5}$ in the UK. This limits the extent to which long-term average concentrations can be reduced by UK action alone.

Figure 4 shows maps of the impacts on 2010 surface $PM_{2.5}$ for 30% reductions in UK terrestrial emissions of each of NH_3, NO_x, SO_x, VOC and primary $PM_{2.5}$. The effect of these emissions reductions on the three measures of UK-average surface concentrations of $PM_{2.5}$ are illustrated in Figure 5, based on the data given in Table 1. The principal observations from the two figures are that $PM_{2.5}$ levels in the UK do not show strong responses to UK-only reductions in emissions of individual components/precursors of $PM_{2.5}$, and that the responses are highly geographically variable. The maximum reduction in $PM_{2.5}$ concentrations (at a 5 km grid resolution) reaches ~0.6 $\mu g\ m^{-3}$ (~6% of the modelled components) in response to a 30% reduction in UK emissions of individual components, and in most locations the reductions in $PM_{2.5}$ concentrations are considerably smaller. This again indicates the influence on $PM_{2.5}$ in the UK (on an annual average basis) from emissions outside of the UK. In the case of the formation of SIA components, it also reflects the non-linearity in the precursor oxidation chemistry and gas-particle phase partitioning that occurs between emission location and receptor location (Harrison et al., 2013;Vieno et al., 2014).

Figure 4 and Figure 5 show that, on average across the UK, the sensitivity of $PM_{2.5}$ concentrations to a 30% reduction in UK emissions of individual contributing components increases in the order VOC, NO_x, SO_x, primary $PM_{2.5}$ and NH_3. The exact order varies slightly with the UK-average measure used (Figure 5). This is due to differences in the spatial patterns of the $PM_{2.5}$ reductions shown in Figure 4 in relation to the distribution of UK population shown in Figure 2.

The 30% reductions in UK VOC emissions gives maximum reductions of ~0.15 µg m^{-3} (1.5%) in $PM_{2.5}$ concentrations in central and northern England and central Scotland (Figure 4e). The 30% reductions in UK NO_x emissions yield around 0.2 µg m^{-3} (3%) reductions in $PM_{2.5}$ over some rural areas (Figure 4c), and generally a maximum of 0.15 µg m^{-3} (1.5%) reductions in $PM_{2.5}$ over other rural areas. An important observation is that reductions of $PM_{2.5}$ over urban centres are smaller (no more than 0.15 µg m^{-3}) than in rural areas for these reductions in NO_x emissions. The 30% reductions in UK SO_x emissions yield up to ~0.45-0.5 µg m^{-3} (5%) reductions in $PM_{2.5}$ in the Trent valley and up to around 0.3-0.35 µg m^{-3} (3%) reductions in $PM_{2.5}$ over large areas of central and northern England and central Scotland (Figure 4d). The locations with greatest sensitivities to the 30% NO_x emissions reductions (Figure 4c) are generally those with the lowest sensitivities to SO_x emissions reductions (Figure 4d). As with the NO_x emissions reductions, the reductions in $PM_{2.5}$ concentrations for reductions in SO_x emissions is not, in general, associated with the major urban areas, except where these also have major SO_x sources in the vicinity (e.g. Trent Valley, West Midlands, Cheshire). This is primarily caused by the spatial distribution of major sources of SO_x emissions. As ~80% of UK SO_x 2010 emissions originate from large point sources (power plants, industrial facilities), which are not located in the heart of urban areas, associated emission reductions have the most profound effects in rural areas. However, the greater sensitivity to SO_x close to large point sources (e.g. coal-fired power plants) may in part be an artefact due to the model assumption that 5% of SO_x emissions are directly in the form of SO_4^{2-}, which may no longer be appropriate for these sources or for models running at relatively high horizontal spatial resolution. The SO_x and NO_x gases compete in their reaction with NH_3 to form particulate ammonium sulphate ($(NH_4)_2SO_4$) or ammonium nitrate (NH_4NO_3). The larger sensitivity of $PM_{2.5}$ formation to NH_3 emissions reductions indicates that NH_3 is the limiting species; whilst the greater sensitivity to SO_x than to NO_x emissions reductions reflects that the reaction between NH_3 and SO_x is fast and essentially irreversible compared with the equilibrium reactions between gaseous NH_3 and NO_x species and NH_4NO_3.

The largest reductions in $PM_{2.5}$ (when weighted towards areas of greatest population) derive from 30% reductions in UK NH_3 and primary $PM_{2.5}$ emissions (Figure 4b and Figure 4f), up to 0.45 µg m^{-3} for NH_3 reductions and greater for primary $PM_{2.5}$ reductions (up to ~6% of modelled $PM_{2.5}$ in both cases). There is a distinct inverse geographic relationship in the $PM_{2.5}$

sensitivity to reductions of these two components. The reductions in NH_3 emissions give greatest $PM_{2.5}$ decreases in agricultural areas, whereas the reductions in primary $PM_{2.5}$ give greatest decreases in the large conurbations and other areas of high population density. The difference in geographical patterns is highlighted more clearly in Figure 6a which shows the data in Figure 4b minus the data in Figure 4f. Blue colours in Figure 6a indicate where reductions in $PM_{2.5}$ from a 30% reduction in NH_3 emissions exceed the reductions in $PM_{2.5}$ from a 30% reduction in primary $PM_{2.5}$ emissions, and vice-versa for red colours. White colours indicate comparable reductions in $PM_{2.5}$ via primary $PM_{2.5}$ or NH_3 emissions reductions. The geographical pattern in $PM_{2.5}$ sensitivity reflects the geographical pattern of the emission sources and the fact that, because of the short atmospheric lifetime of NH_3, UK emissions of NH_3 also generally have short-range influence.

Figure 7 shows the map of annual-average surface concentration of $PM_{2.5}$ estimated for the 2030 CLE emissions projections, and of the difference between the $PM_{2.5}$ concentrations in 2030 and 2010. Surface concentrations of $PM_{2.5}$ over the UK are simulated to reduce by up to 2.8 $\mu g\ m^{-3}$ between 2010 and the 2030 CLE emissions scenario used. The UK-wide reductions in $PM_{2.5}$ between 2010 and 2030 CLE are 1.70, 2.24 and 2.42 $\mu g\ m^{-3}$ for the area-weighted, population-weighted and AEI summary measures, respectively. The impacts on surface $PM_{2.5}$ in 2030 of additional 30% reductions applied to UK-only terrestrial emissions of each of NH_3, NO_x, SO_x, VOC and primary $PM_{2.5}$ individually are shown in Figure 8. Figure 5 illustrates the quantitative effect of these further emissions reductions against the 2030 CLE scenario on the three summary measures of UK-average surface concentrations of $PM_{2.5}$.

The maps in Figure 8 show qualitatively very similar findings to their equivalent maps in Figure 4. In 2030, UK $PM_{2.5}$ is projected to remain more sensitive to reductions in UK emissions of NH_3 and primary $PM_{2.5}$ than to reductions in UK SO_x and NO_x; and, from a population-weighted perspective, to be relatively more sensitive to further primary $PM_{2.5}$ and NH_3 emissions reductions, particularly to primary $PM_{2.5}$ emissions reductions, than was the case for the 2010 simulations (Figure 5). For the 2030 simulations, additional 30% reductions in UK primary $PM_{2.5}$ or NH_3 emissions yield reductions in $PM_{2.5}$ of up to 0.5 $\mu g\ m^{-3}$ or 0.25 $\mu g\ m^{-3}$, respectively (Figure 8), whilst in 2010 additional 30% reductions in primary $PM_{2.5}$ or NH_3 emissions yield reductions in $PM_{2.5}$ of up to 0.6 $\mu g\ m^{-3}$ or 0.45 $\mu g\ m^{-3}$, respectively (Figure 4).

The 2030 results again emphasise a geographic pattern of greatest sensitivity of $PM_{2.5}$ to reductions in the areas of high population density. Figure 6b plots the difference in response to the NH_3 and primary $PM_{2.5}$ emissions reductions in 2030, analogous to the plot in Figure 6a for the 2010 sensitivities. Figure 6b clearly emphasises that for this projection for 2030, UK $PM_{2.5}$ is relatively even more sensitive to further reductions in UK primary $PM_{2.5}$ emissions compared with further reductions in UK NH_3 emissions, particularly in populated areas, than is the case for 2010; albeit that the additional absolute reductions in $PM_{2.5}$ for a given percentage of emissions reductions is smaller in 2030 than in 2010 (Figure 5) because of the general decline in emissions across Europe during this period for this scenario.

4 Discussion

Simulations were undertaken for both 2010 and a 2030 scenario to investigate whether conclusions on effectiveness of potential UK mitigation differ between the two time points. It is recognised that reductions in emissions of primary $PM_{2.5}$ and precursor gases from many anthropogenic sources are already anticipated going forward under current legislation, so it is important to know, for a future policy perspective, the anticipated sensitivities of UK $PM_{2.5}$ to additional UK emission reductions in the future.

The simulations for both 2010 and 2030 CLE show that if the focus is on the reduction of spatially-averaged $PM_{2.5}$ concentrations then the most effective UK control, via an individual component, is achieved through reduction of UK emissions of NH_3, as shown in Figure 5. However, the conclusion is different when considering population-weighted $PM_{2.5}$ reductions for the mitigation of human health effects. For a full population weighting across all 5 km × 5 km model grids, reductions in UK primary $PM_{2.5}$ emissions are almost as effective as reductions in UK NH_3 emissions for the 2010 simulations, but primary $PM_{2.5}$ emissions reductions are simulated to be the most effective additional control in the 2030 CLE future (Figure 5). Emphasis on population weighting also increases the sensitivities of $PM_{2.5}$ to reductions in NO_x emissions in both 2010 and 2030 CLE because a major source of NO_x is road traffic whose emissions are associated with where population live. On the other hand, the sensitivity of $PM_{2.5}$ to further reductions in UK SO_x emissions is markedly lower in 2030 than in 2010 because of the large reductions in SO_x emissions already implemented under the CLE scenario. It is also recognised that reductions in NO_x and VOC emissions have the potential to deliver health benefits separately from their contribution to reduction in $PM_{2.5}$ through reductions in population exposure to surface NO_2 and O_3.

An important observation is that the effectiveness of emissions reductions on $PM_{2.5}$ using a population weighting for the quantification differs between evaluation via full nation-wide gridded population-weighting or via use of data only at the locations used to derive the AEI. Quantification through the AEI puts greater emphasis on the effectiveness of primary $PM_{2.5}$ emissions reduction, and on NO_x emissions reductions, (Figure 5) because the monitor locations contributing to the AEI are sited in the largest cities and towns where emissions of primary $PM_{2.5}$ and NO_x are prevalent. Based on the AEI, control of primary $PM_{2.5}$ is the most effective individual component in 2010 as well as in 2030 CLE. These observations are pertinent given that the AEI is the air quality metric for $PM_{2.5}$.

Analyses from the EUCAARI study in Kulmala et al. (2011) and a more recent European study in Megaritis et al. (2013) both suggest that reducing NH_3 emissions is the most effective way to reduce $PM_{2.5}$ under present-day conditions. Whilst the current study also emphasises the sensitivity of $PM_{2.5}$ to NH_3 emissions reductions, it also emphasises that, for the UK, a sensitivity to primary $PM_{2.5}$ emissions reductions is at least as great as for NH_3 when considering population-weighting of $PM_{2.5}$ concentrations, both currently and for a future CLE scenario. In fact the sensitivity to primary $PM_{2.5}$ emissions may be underestimated by the simulations because of dilution of primary $PM_{2.5}$ emissions into the 5 km × 5 km grid resolution of the model. It has been calculated that a 1:1 relationship between UK primary $PM_{2.5}$ emissions reductions and the reduction in the primary $PM_{2.5}$ component of the UK 2010 AEI would lead to a reduction in the 2010 AEI of 0.8 $\mu g\ m^{-3}$ (AQEG, 2015), compared with the 0.37 $\mu g\ m^{-3}$ derived from the model simulations in this work (Table 1). Even so, the total impact of 30% reductions in UK emissions of all the components/precursors listed in Table 1 on the 2010 baseline, is only of comparable magnitude (1.2 $\mu g\ m^{-3}$) to the 15% (or 1.3 $\mu g\ m^{-3}$) reduction required in the UK AEI by 2020. However, reductions in these emissions from outside the UK will also contribute to reducing the UK $PM_{2.5}$ AEI. Conversely, reductions of emissions in the UK will also yield benefits for surface $PM_{2.5}$ concentrations elsewhere in Europe. The country-to-country source-receptor matrices developed by EMEP MSC-W at the 50 km resolution indicate that reductions in the UK of the same primary and precursor species considered in this work would (for 2011 emissions) lead to reductions in $PM_{2.5}$ in neighbouring countries up to about one-third the magnitude of the $PM_{2.5}$ reductions in the UK (Fagerli et al., 2014). Reductions of emissions in the UK would also lead to other benefits outside the UK on, for example, NO_2 and O_3 exposure and on N and S deposition.

Although the model used in this study is widely applied across Europe for air quality policy development (Fagerli et al., 2014), the data presented here are from simulations from a single model. The model simulations of the effect of inorganic precursor gases on the secondary inorganic $PM_{2.5}$ are dependent on accurate representation of the relevant chemistry and phase partitioning. It is possible that the SIA representation in the EMEP4UK model may underestimate the nitrate in the $PM_{2.5}$ size fraction, and hence downplay somewhat the sensitivity of $PM_{2.5}$ to NO_x emissions reductions. In addition, not explicitly calculating the uptake of HNO_3 by mineral dust may reduce the NO_3^- changes due to NO_x emissions reduction. However, the EMEP4UK particle sulphate, nitrate and ammonium concentrations all compare well with the multi-year time series of measurements of these components at ~30 sites across the UK in the Acid Gas and Aerosol Network (AGANet) and National Ammonia Monitoring Network (NAMN) (Vieno et al., 2014). Variation in particle-bound water may also impact on the exact $PM_{2.5}$ mass sensitivities associated with inorganic precursor gas emissions reductions.

Inter-annual variability in meteorology may also have an influence, in particular in determining the balance in any year between $PM_{2.5}$ in the UK derived from UK emissions and that derived from emissions outside the UK (Vieno et al., 2014). However, whilst the precise quantitative sensitivities of annual average $PM_{2.5}$ to emissions reductions will be subject to inter-annual meteorological variability, it is anticipated that the broad findings of this study will hold.

The interpretation of the modelling results has been undertaken from the perspective that reduction in all anthropogenically-derived components of $PM_{2.5}$ is equally important. This remains the current position for the EU legislation that sets limits and targets for concentrations of $PM_{2.5}$ (Heal et al., 2012); i.e. no consideration is given to the potential different toxicity to human health of different components of $PM_{2.5}$. The UK Committee on the Medical Effects of Air Pollutants has also recently concluded that reductions in concentrations of both primary and secondary particles are likely to benefit public health (COMEAP, 2015). Nevertheless, although not conclusive, there is evidence that traffic-related sources of PM, or combustion sources more generally, are particularly associated with adverse health outcomes (Grahame and Schlesinger, 2007, 2010;Janssen et al., 2011;Stanek et al., 2011;WHO, 2013;Grahame et al., 2014). The possibility that primary $PM_{2.5}$ is more toxic per unit mass than secondary $PM_{2.5}$, places greater emphasis on the finding from this work on the effectiveness of reductions in emissions of primary $PM_{2.5}$. Interpretation of the modelling results has also not considered the relative costs or feasibilities of implementing further reductions in the emissions of the individual precursors and components investigated.

Finally, it should be remembered that $PM_{2.5}$ has impacts other than on human health, although reduction in urban background concentrations through the $PM_{2.5}$ AEI is in legislation. Measures taken in the UK to reduce concentrations of ambient $PM_{2.5}$ and of precursor gases, both within and outside of areas of population, will have multiple co-benefits on human health, N and S deposition, ozone formation and radiative forcing, not just in the UK but elsewhere.

5 Conclusions

The sensitivity of annual-average surface concentrations of $PM_{2.5}$ across the UK to reductions in UK terrestrial anthropogenic emissions in primary $PM_{2.5}$, NH_3, NO_x, SO_x and non-methane VOC was investigated using the EMEP4UK atmospheric chemistry transport model for 2010 and for a 2030 current legislation scenario that includes projected pan-European emission changes. In general, the sensitivity of modelled concentrations to UK-only mitigation is rather small. A 30% reduction in UK emissions of any one of the above listed PM components yields (for the 2010 simulation) a maximum reduction in $PM_{2.5}$ concentrations in any given location of ~0.6 $\mu g\ m^{-3}$ (equivalent to ~6% of the total modelled $PM_{2.5}$ mass concentration). On average across the UK, the sensitivity of $PM_{2.5}$ concentrations to a 30% reduction in UK emissions of individual contributing components, for both the 2010 and 2030 CLE baselines, increases in the order NMVOC, NO_x, SO_x, NH_3 and primary $PM_{2.5}$, but there are strong spatial differences in the $PM_{2.5}$ sensitivities across the UK. Consequently, the sensitivity of $PM_{2.5}$ to individual component emissions reductions varies between area and population weighting. Reductions in NH_3 have the greatest area-weighted effect on $PM_{2.5}$. A full UK population weighting places greater emphasis on reductions of primary $PM_{2.5}$ emissions, which is simulated to be the most effective single-component control on $PM_{2.5}$ for the 2030 scenario. An important observation is that weighting corresponding to the Average Exposure Indicator metric (using data from the 45 model grids containing a monitor whose measurements are used to calculate the UK AEI) further increases the emphasis on the effectiveness of primary $PM_{2.5}$ emissions reductions (and of NO_x emissions reductions) relative to the effectiveness of NH_3 emissions reductions. Reductions in primary $PM_{2.5}$ has the largest impact on the AEI in 2010 as well as the 2030 CLE scenario. The summation of the reductions to the UK $PM_{2.5}$ AEI of the 30% reductions in UK emissions of primary $PM_{2.5}$ and of NH_3, SO_x, NO_x and VOC totals ~1.2 µg m^{-3} and ~0.8 µg m^{-3} with respect to the 2010 and 2030 CLE baselines, respectively.

Acknowledgements

This work is funded jointly by the UK Department for the Environment, Food and Rural Affairs (Defra), the NERC Centre for Ecology and Hydrology (CEH), the EMEP programme under the UNECE LRTAP Convention, the Norwegian Meteorological Institute (Met.No) and the European Union projects NitroEurope IP and ÉCLAIRE.

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Table:

Table 1: EMEP4UK-modelled estimates of the impact of 30% UK terrestrial emissions reductions on three measures of UK-average surface concentrations of $PM_{2.5}$ (µg m^{-3}): (i) the average of the model grids containing the 45 monitors used to calculate the UK $PM_{2.5}$ Average Exposure Indicator (AEI), (ii) the population-weighted average, and (iii) the area-weighted (i.e. geographical) average, for 2010, and for 2030 under a CLE emission scenario (using 2010 meteorology). For context, the modelled reductions in the baselines between 2010 and 2030 CLE for the three measures of UK-average $PM_{2.5}$ are 2.42, 2.24, and 1.70 µg m^{-3}, respectively.

Emissions reduced	**AEI**		**Population-weighted**		**Area-weighted**	
	2010	**2030 CLE**	**2010**	**2030 CLE**	**2010**	**2030 CLE**
Primary $PM_{2.5}$	0.37	0.29	0.31	0.24	0.16	0.13
NH_3	0.35	0.19	0.34	0.19	0.28	0.16
SO_x	0.27	0.15	0.26	0.15	0.19	0.11
NO_x	0.10	0.14	0.10	0.15	0.11	0.13
VOC	0.08	0.05	0.08	0.05	0.07	0.03
Total	**1.17**	**0.82**	**1.10**	**0.77**	**0.82**	**0.57**

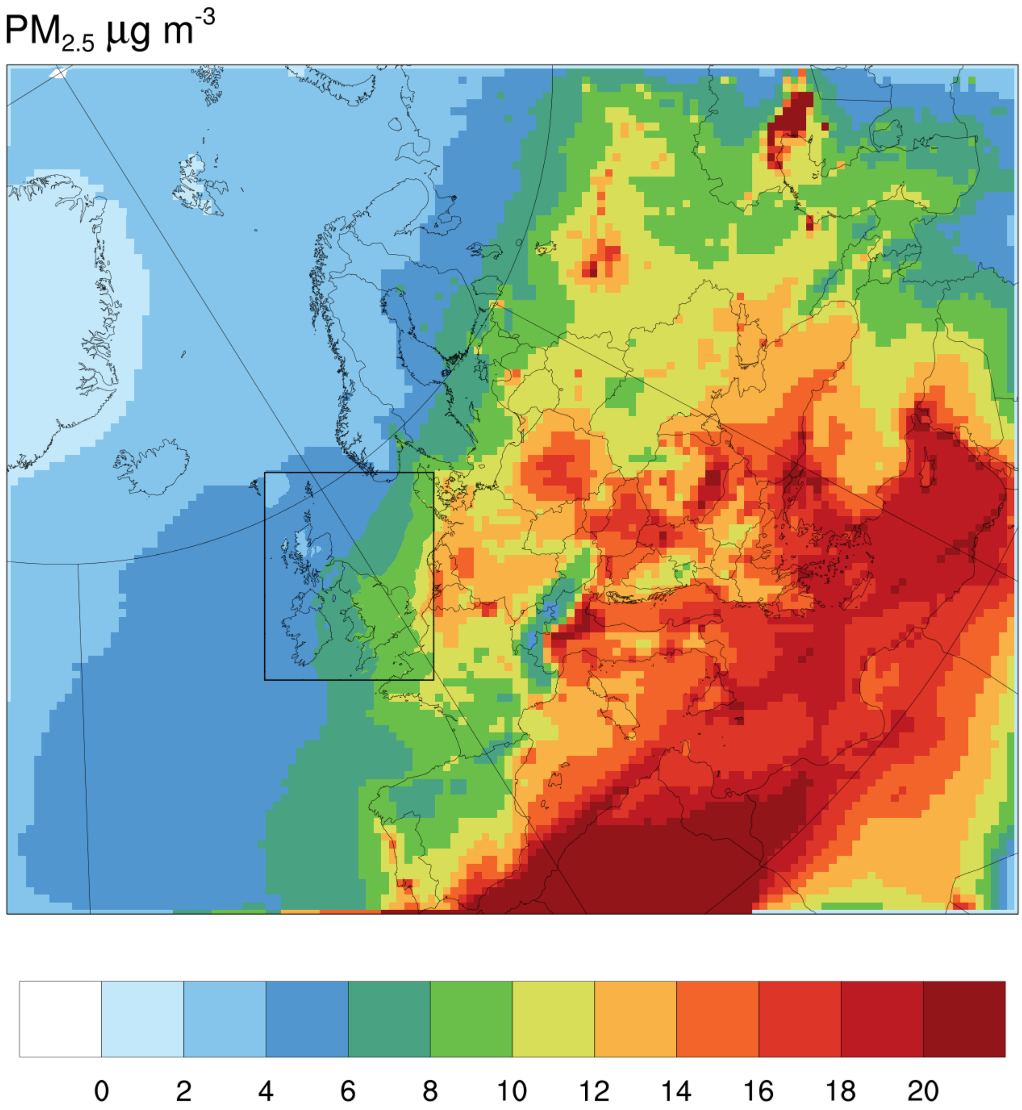

Figure 1: 2010 EMEP4UK annual-average surface concentrations of PM$_{2.5}$ (µg m^{-3}) at 50 km × 50 km horizontal resolution for the European model domain, and at 5 km × 5 km horizontal resolution for the nested British Isles domain (black box).

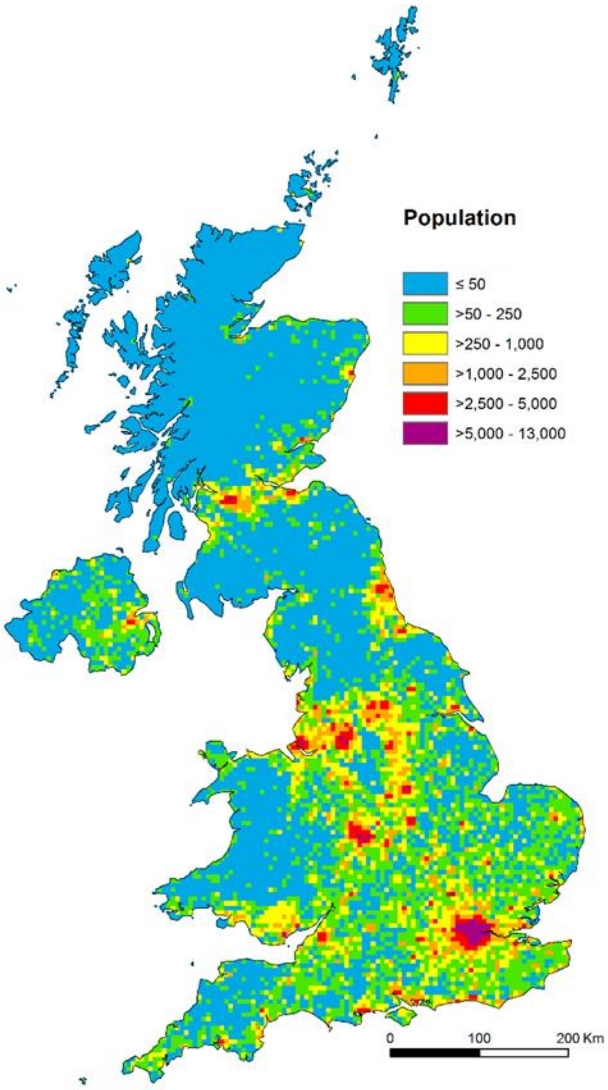

Figure 2: Gridded UK population density based on the UK census at the 5 km × 5 km grid spatial resolution. Units are population km^{-2}.

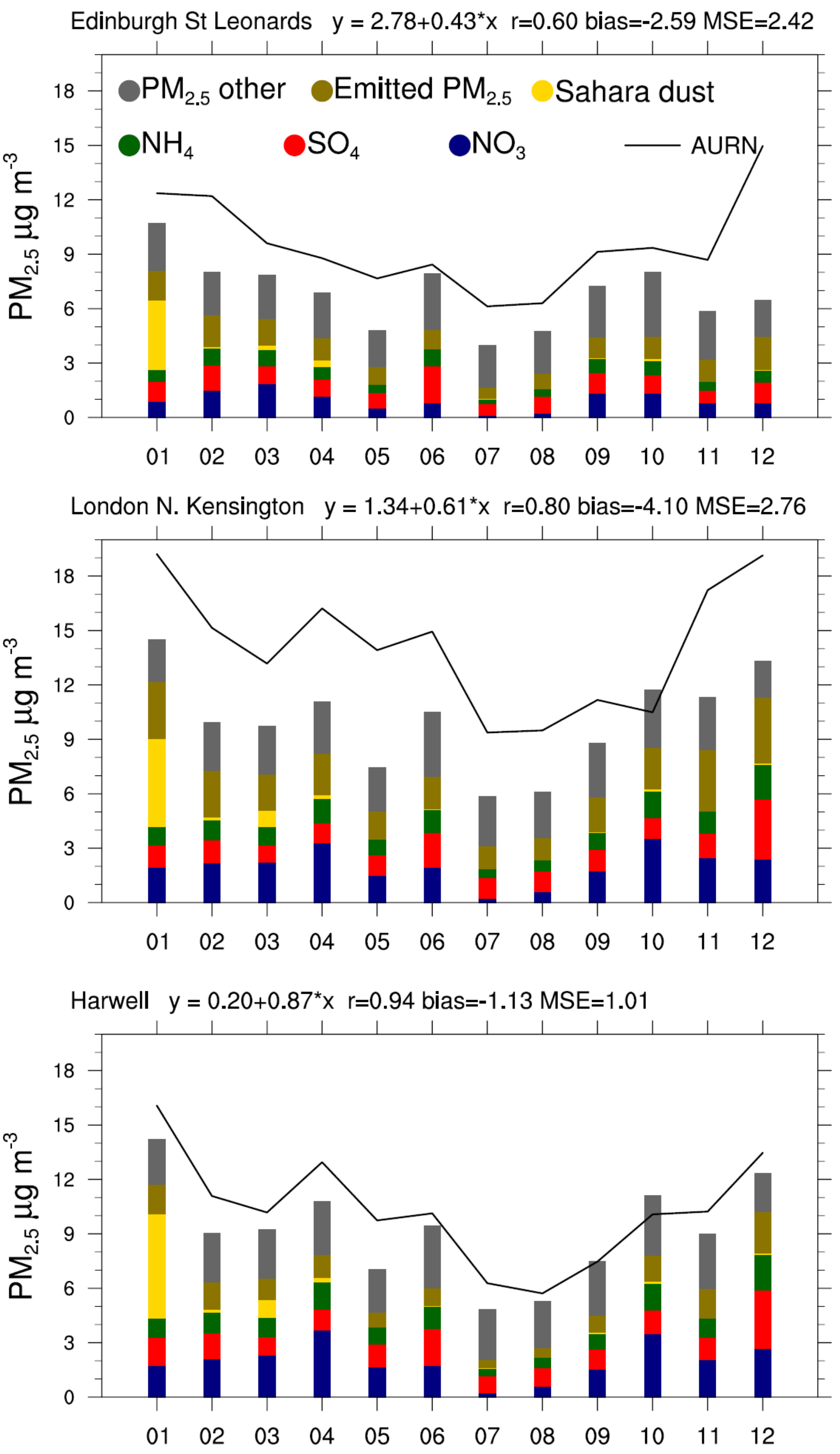

Figure 3: 2010 monthly-averaged EMEP4UK simulated $PM_{2.5}$ components and total $PM_{2.5}$ observations by TEOM-FDMS at the Edinburgh St. Leonards, London North Kensington and Harwell UK national network (AURN) monitoring sites. Both the modelled and observed data are averaged from hourly values. The linear regression between observation and model is also shown at the top of each panel, along with the correlation coefficient, r, bias and mean square error.

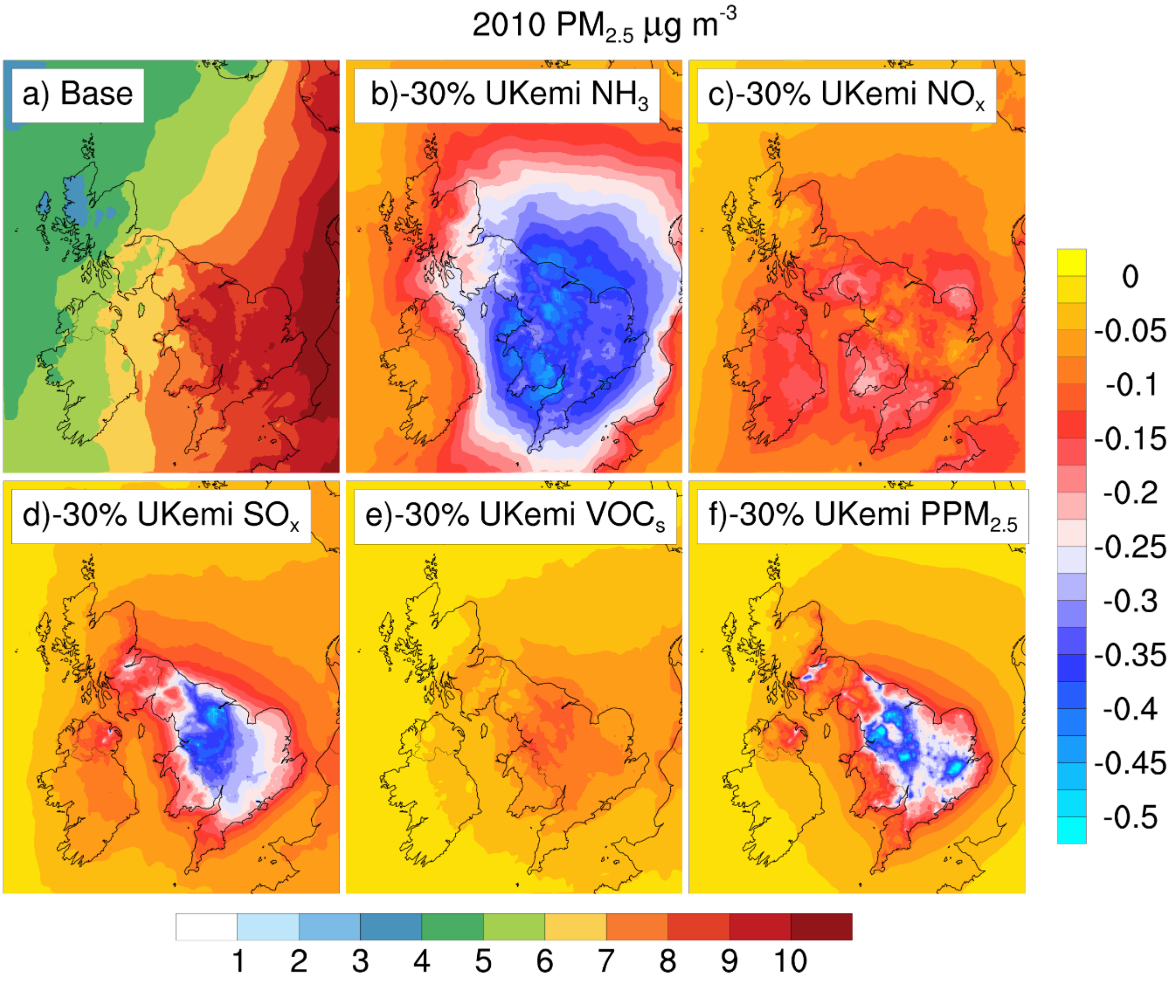

Figure 4: Model simulations of the impact of 30% UK emissions reductions on annual-average surface concentration of $PM_{2.5}$. Panel (a) 2010 base-case scenario, no emissions reduction (bottom colour scale); remaining panels, the change in annual-average $PM_{2.5}$ for 30% UK emissions reductions in (b) NH_3, (c) NO_x, (d) SO_x, (e) VOC, and (f) primary $PM_{2.5}$ (right colour scale). All units are µg m^{-3}.

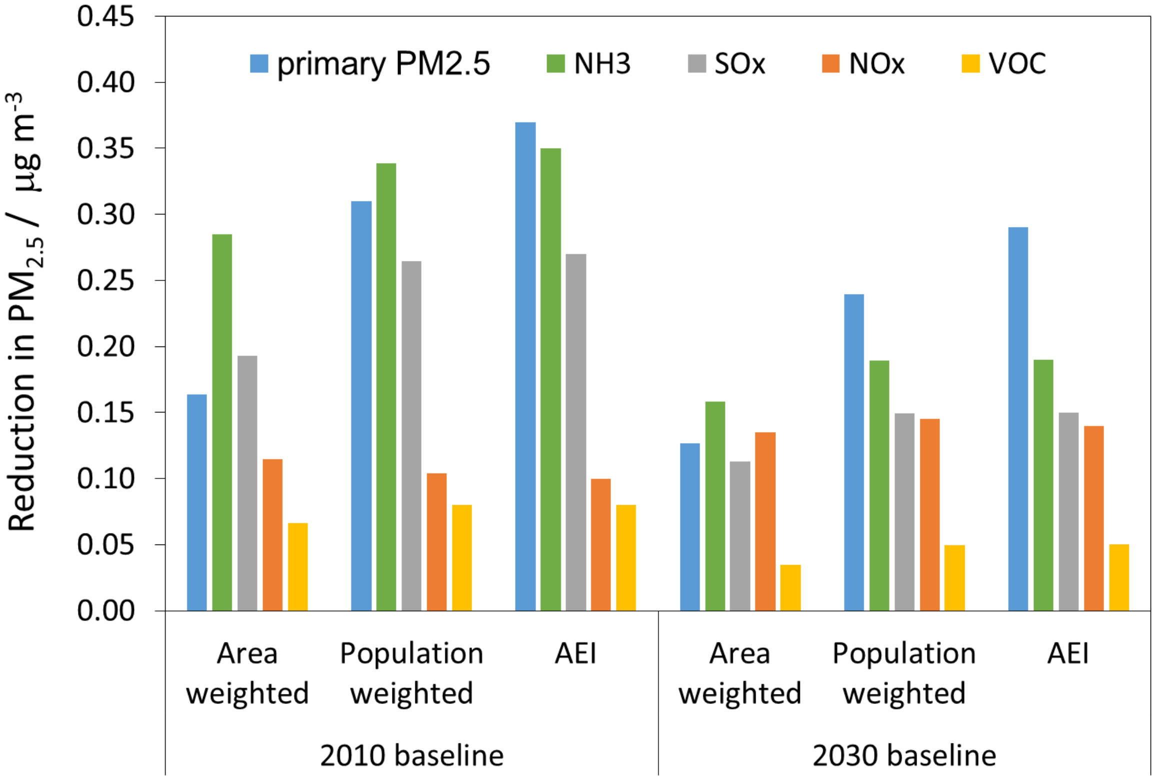

Figure 5: The impact of 30% UK terrestrial emissions reductions in primary $PM_{2.5}$, NH_3, SO_x, NO_x, and VOC (individually) on three measures of UK-average surface concentrations of $PM_{2.5}$: area weighted; population weighted; and the average for the 45 model grids containing the monitors used to calculate the UK $PM_{2.5}$ Average Exposure Indicator (AEI). Data are shown for simulations for 2010, and for 2030 under a CLE emission scenario (using 2010 meteorology).

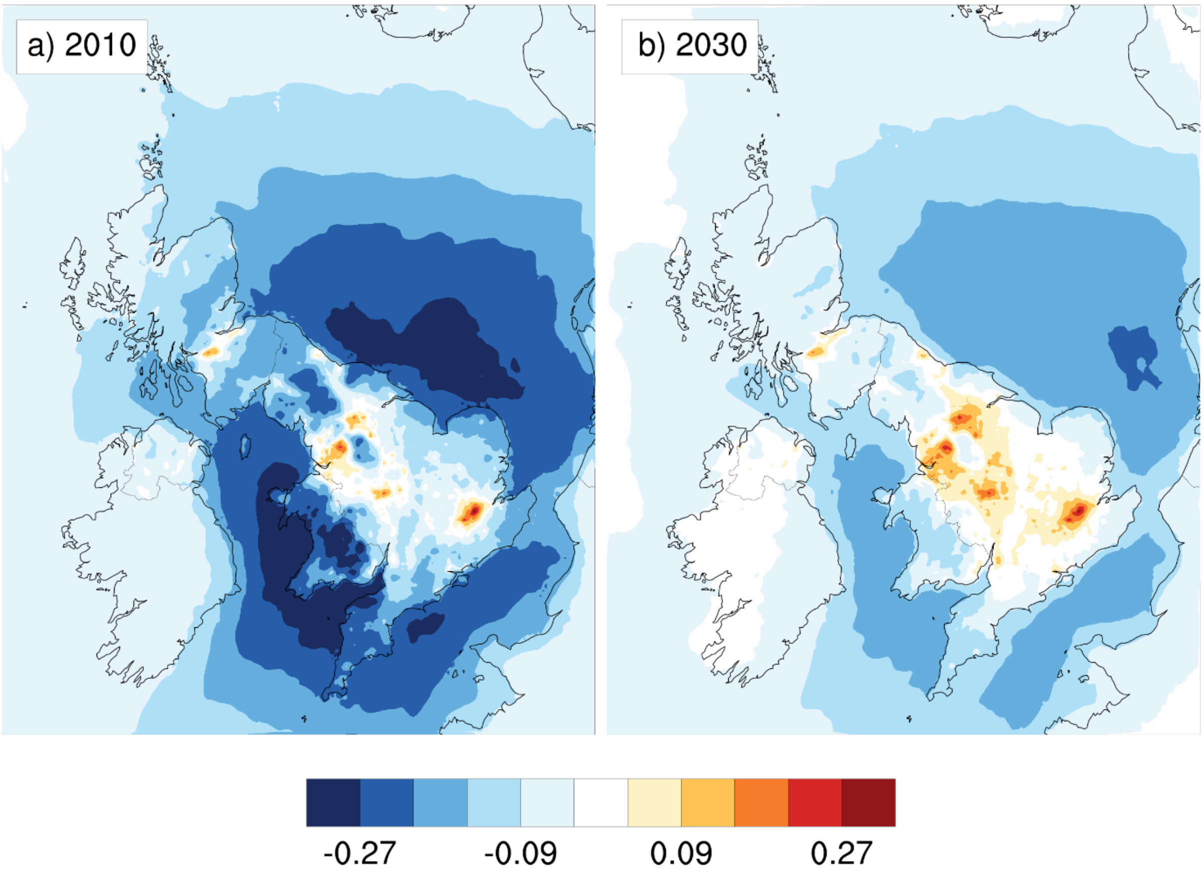

Figure 6: The difference between changes in simulated annual-average $PM_{2.5}$ (µg m^{-3}) for 30% reductions in UK NH_3 emissions reduction and for 30% reductions in UK primary $PM_{2.5}$ emissions reduction: (a) for the year 2010 (i.e. the data in Figure 4b minus the data in Figure 4f); and (b) for the year 2030 (i.e. the data in Figure 8b minus the data in Figure 8f). Blue colours indicate where reductions in $PM_{2.5}$ for 30% reduction in NH_3 emissions exceed the reductions in $PM_{2.5}$ for 30% reduction in primary $PM_{2.5}$ emissions, and vice versa for the red colours. The same meteorological year 2010 was used.

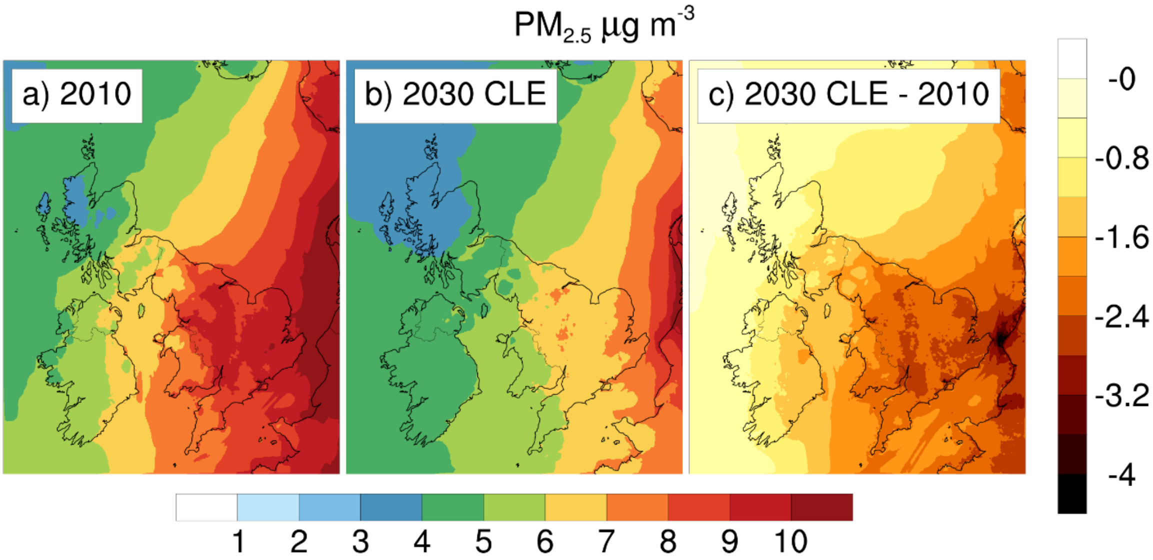

Figure 7: EMEP4UK annual-average surface concentration of $PM_{2.5}$ (µg m^{-3}) for a) 2010 emissions, and b) 2030 CLE emissions projection (bottom colour scale), and c) the difference 2030 CLE – 2010 CLE (right colour scale). The same meteorological year 2010 was used.

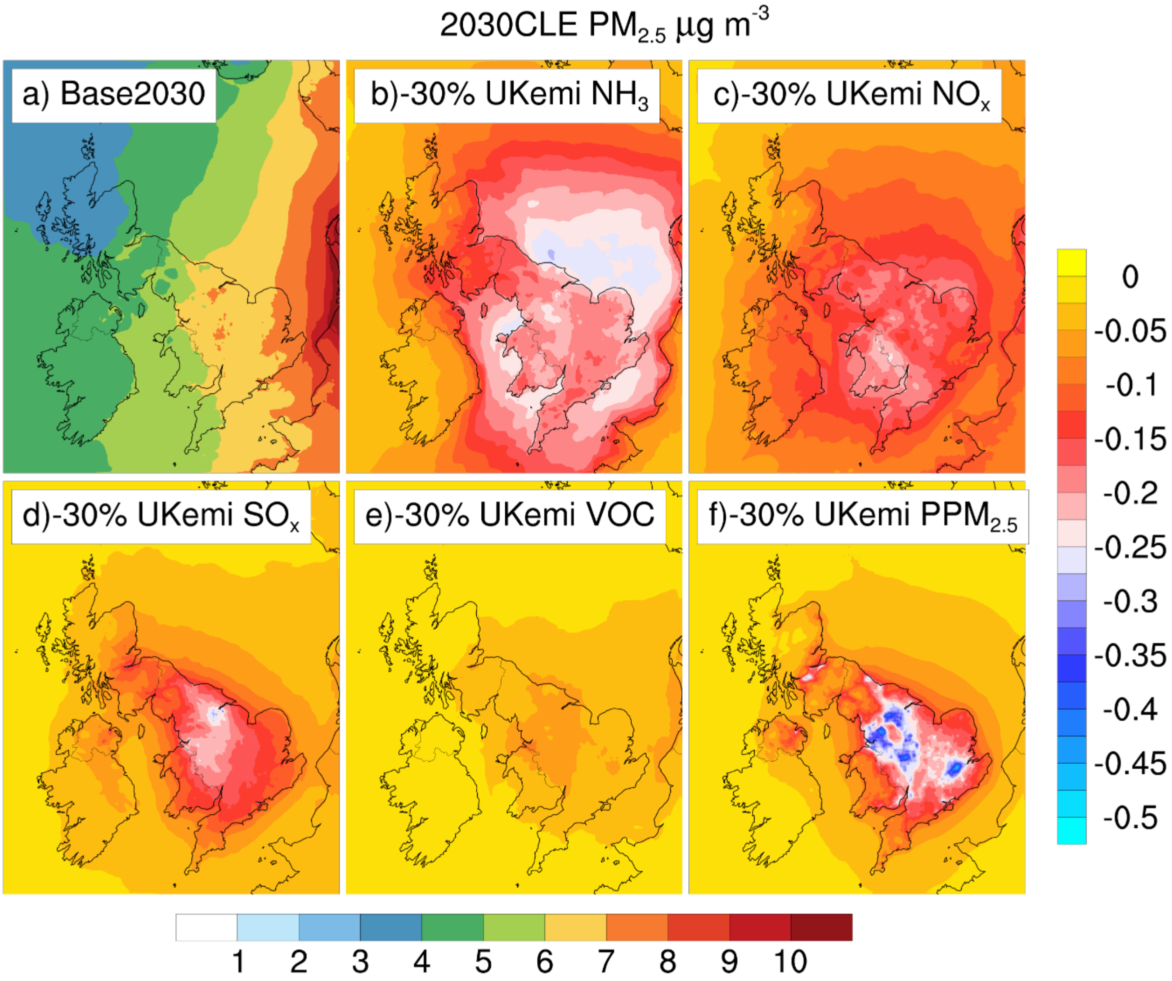

Figure 8: Model simulations of impact of 30% UK emissions reductions on annual-average surface concentration of $PM_{2.5}$ for a future scenario (with 2010 meteorology). Panel (a), 2030 CLE scenario, no emissions reduction (bottom colour scale); remaining panels, the change in annual-average $PM_{2.5}$ for 30% UK emissions reductions in (b) NH_3, (c) NO_x, (d) SO_x, (e) VOC, and (f) primary $PM_{2.5}$ (right colour scale). All units are µg m^{-3}.